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LLNL-JRNL-411666

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March 30, 2009

Journal of Radioanalytical and Nuclear Chemistry

Log No. 272

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ABSTRACT

This work provides an assessment of suitable forensic indicators that may be measured by portable mass spectrometry systems. Traditional assessments of nuclear fuel cycle manipulation or other nuclear activities often depend upon analyses of uranium and plutonium isotopes in the nuclear fuel. Any entity engaging in shortened fuel cycle activity will recover U and Pu during reprocessing. Fission, capture, and activation products are less valuable and generally regarded as waste products. This work determined isotopic ratios that distinguish nuclear weapons and shortened nuclear fuel cycles from commercial nuclear reactors. Modeling of fuel cycles was conducted via ORIGEN-S, MCNPX, and through custom calculations.

INTRODUCTION

This work provides an assessment of suitable forensic indicators that may be measured by portable mass spectrometry systems. Measurement by mass spectrometry is required to reduce the time lag between sample collection and analysis experienced by decay counting. Isotope ratios within individual elements are best for analysis because any chemical process during transport or ionization affects all isotopes equally. Traditional assessments of nuclear fuel cycle manipulation or other nuclear activities often depend upon analyses of uranium and plutonium isotopes in the nuclear fuel.[1] Any entity engaging in shortened fuel cycle activity will recover U and Pu during reprocessing. Fission, capture, and activation products are less valuable and generally regarded as waste products. This work determined isotopic ratios that distinguish nuclear weapons and shortened nuclear fuel cycles from commercial nuclear reactors. The bulk of this paper deals with the modeling of the reactor test cases for preliminary assessment of possible isotopic ratios. Mass spectrometric methods will be assessed to determine the detection limits of the isotopic ratios identified in the modeling aspect of this work. The isotope ratio lists generated will be independent of current measurement capabilities and can serve as design criteria for future field deployable systems.

There are many sources for radionuclides in our environment which can include natural sources, the commercial nuclear industry, nuclear weapons, and the medical industry.[1] Often times, the source of the radionuclide may be determined through just identification of the radionuclide. If radionuclides are produced through different sources, the identification of the source is complex. In order to ascertain a specific source for attribution, radionuclide ratios are often employed.

Production yields of radionuclides from fission are a function of many variables including: the fissile material, the energy spectrum of the neutron flux, the magnitude of the neutron flux, and the duration of the irradiation. As a result, the ratios of certain radionuclides are highly dependent of these variables and may be utilized to distinguish between radionuclides produced from nuclear weapons, medical waste, short nuclear fuel cycles (*e.g.* ^{239}Pu production fuel cycles), and long nuclear fuel cycles (*e.g.*, commercial nuclear fuel cycles). While the above is easily stated, the difficult part is to determine which radionuclide ratios should be utilized for best forensic value.

As an added complication, nuclear debris taken for forensic analysis often does not come directly from the source. There is often some type of transport process or other process that may alter the sample composition. Chemical fractionation can occur and may significantly alter ratios of radionuclides of different elements. To mitigate this problem, it is best to examine isotopic ratios of individual elements since these ratios will be largely unaltered by chemical processes.[1, 2, 3]

METHOD

This work will investigate isotopic ratios that distinguish nuclear weapons and shortened nuclear fuel cycles from commercial nuclear reactors. The difference between ratio values will be determined via Equation 1.

$$R = \frac{\left({}^{A}_{Z}X\right)_{weapon} \Big/ \left({}^{B}_{Z}X\right)_{weapon}}{\left({}^{A}_{Z}X\right)_{reactor} \Big/ \left({}^{B}_{Z}X\right)_{reactor}} \qquad (1)$$

where

$\left({}^{A}_{Z}X\right)_{reactor}$ is the concentration of isotope A produced in a power reactor

$\left({}^{B}_{Z}X\right)_{reactor}$ is the concentration of isotope B produced in a power reactor

$\left({}^{A}_{Z}X\right)_{weapon}$ is the concentration of isotope A produced in a weapon/short burn

$\left({}^{B}_{Z}X\right)_{weapon}$ is the concentration of isotope B produced in a weapon/short burn

Modeling was done using ORNL isotope generation and depletion code ORIGEN-ARP and LANL MCNPX. Three reactor types were modeled within ORIGEN: BWR, PWR, and CANDU. For each reactor type, an appropriate normal burnup length was chosen as well as 5% of a normal length. The short irradiation time is used to account for those fuel cycles focused on ^{239}Pu production rather than nuclear power in a commercial setting. From these tests, all fission products and actinides in the ORIGEN-ARP library were tracked, including meta-stable states, as any field deployable mass spectrometry unit would likely be unable to distinguish the two. From the data acquired by the ORIGEN simulations, each possible isotopic ratio was computed for each element present in the output. An algorithm was set up to calculate each unique combination of these elements given that the value of the mass was above some threshold limit dictated by the minimum detectable concentration quoted by common mass spectrometry units. In Table 1, the input parameters are given for each reactor type modeled. Considerations must also be taken to exclude any isotopes that may have half-lives that are unacceptable given the time scales needed to acquire a sample in the field. For the purposes of our test, a decay case was included in the ORIGEN simulation that allowed the fuel isotopics to undergo standard decay for a period that would be suitable for field applications.[4]

Other simulations were completed in MCNPX to model the isotope production from a bare sphere fast reactor of high enrichment. While this is a rather simple model, from an isotope creation/depletion standpoint, this is a decent approximation to what is produced in a nuclear weapon. In MCNPX, isotope tracking was completed for a large set similar to the ORIGEN set using the new predictor-corrector enhanced burnup/depletion (BURN card).[5] Other benefits of using MCNPX for this calculation include the modeling of a fast neutron energy spectrum for the flux given this critical bare isolated sphere configuration of ^{235}U. This data was compared to normal BWR fuel cycle data obtained from the ORIGEN calculations. For the purposes and limits of mass spectrometry, the values for our ratio metric are expected to show at least two orders of magnitude difference between the various fuel cycle cases.

RESULTS

In our analysis, we have found that there are many ratios in the fission product regime that differ by orders of magnitude between short and normal reactor operations. Figures 1 through 3 show the results of this calculation for a PWR, BWR, and CANDU, respectively. From these figures, there are a number of elements that have ratios among their produced isotopes that meet the criteria of having a factors larger than 1000 and even 10000 in some cases. The 100-times criterion was arbitrarily chosen for this study, where lower and more sensitive values may be used given the limitations of the sampling apparatus. Table 2 shows the results for the PWR test cases. The figures are showing peaks across all elements in which the data point indicates the largest found ratio value in the results of the data analysis. The largest ratio value is not necessarily the largest in magnitude but rather the largest distance from unity, as ratios that are very small are also of interest. In addition to these fission products, we can see some transuranic signatures as well. These include the commonly used plutonium ratio as well as americium and curium. The next set of results show the ratio values for the critical bare sphere case that was modeled in MCNPX. Figure 4 shows peaks for the same max ratios along any element vector and the most abundant are those for tin, xenon, and cesium. Some key ratios from this run are listed in Table 2, as well. It is important to note that just values for the PWR are shown. BWR and CANDU runs identified similar isotopic ratios. Figure 5 shows the results for a single element, plutonium, and where the maximum occurs within that array for the PWR simulation in ORIGEN. The results show that certain elements do possess values that exceed the two orders of magnitude requirement for decent mass spectrometry analysis.

CONCLUSIONS

The data has shown that there exist a number of possible isotopic ratios in the family of fission and activation products. Certain elements have shown ratios over 1000 such as those for elements Sr, Cd, Xe, Ba, and Sm, shown in Table 2. These values were for ratios between the long and short irradiation cases for typical light water reactors and CANDU designs. With these signatures, aspects of the reactor's operating procedures could be determined. With further analysis into the sensitivity of these ratios to reactor power, burnup, decay time, and fuel composition, there are possibly more dependencies that could be found, allowing for better on-site sampling procedures for the mass spectrometry units.

With the MCNPX tests that modeled the isotope production in a fast fission spectrum in a critical bare sphere, further modeling is required to correctly account for the time-dependent nature of the isotopics given some decay time after the event. These tests have shown that there exist possible signatures using Kr, Zr, Ce, and Nd.

Further work will be conducted to further characterize detection limits of various mass spectrometry units. Additional work will also be done to better simulate the isotope creation/depletion using more robust codes such as MonteBurns as a bridge between MCNPX and ORIGEN. The isotope library in this framework is much larger and will contain meta-stable states that MCNPX cannot offer alone.

ACKNOWLEDGEMENTS

This work is supported by the Defense Threat reduction Agency under HDTRA1-08-0032. Work performed in part under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

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LIST OF FIGURES

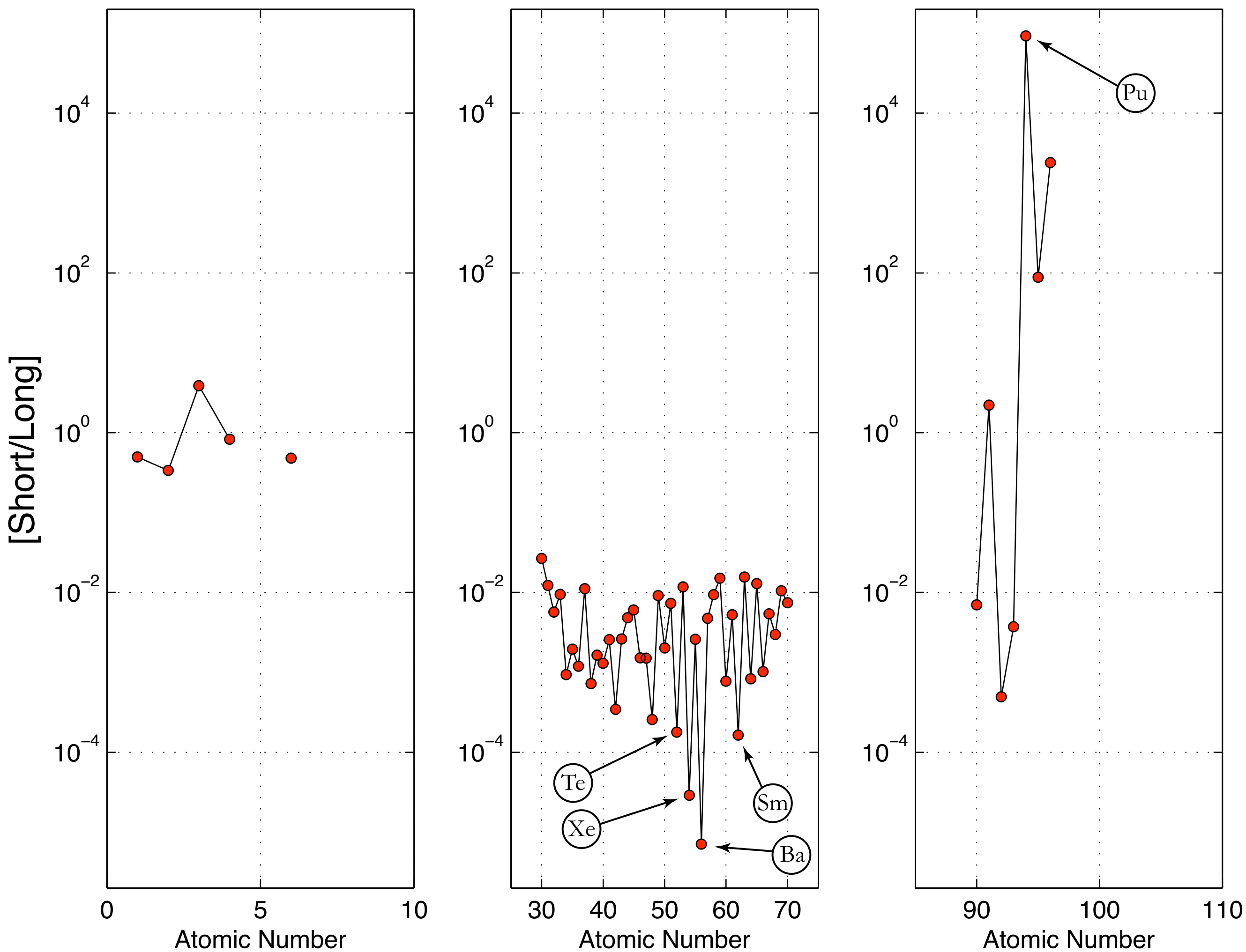
TABLE 1

Type	Enrichment	Irradiation Time (days)	Power Level (MW)
BWR	3%	1460	3579
PWR	4%	1650	3411
CANDU	0.711%	900	2180

TABLE 2. Values for the ratio-of-ratios for possible signature isotopic ratios.

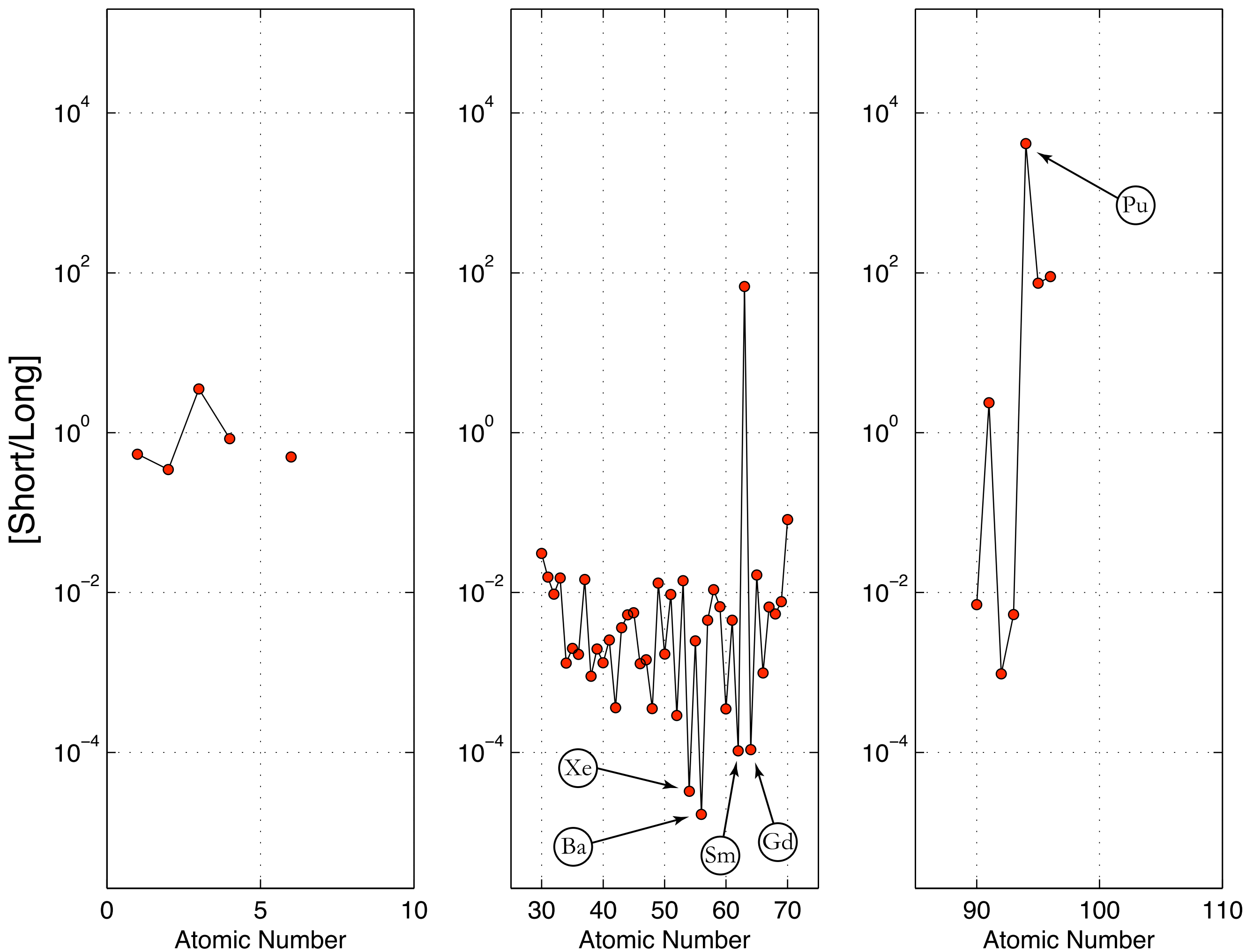
	ORIGEN - PWR (Short Burn/Long Burn)	MCNPX PWR Long Burn vs. Bare Sphere
^{86}Sr to ^{89}Sr	9.898×10^{-4}	-
^{108}Cd to ^{113}Cd	8.184×10^{-4}	-
^{129}Xe to ^{131}Xe	5.996×10^{-4}	-
^{129}Xe to ^{134}Xe	8.112×10^{-4}	-
^{129}Xe to ^{136}Xe	8.796×10^{-4}	-
^{135}Ba to ^{138}Ba	1.972×10^{-4}	-
^{146}Sm to ^{149}Sm	1.635×10^{-4}	-
^{146}Sm to ^{151}Sm	5.245×10^{-4}	-
^{84}Kr to ^{86}Kr	-	2.121×10^{-3}
^{94}Zr to ^{96}Zr	-	6.728×10^{-3}
^{142}Ce to ^{144}Ce	-	9.930×10^{-3}
^{148}Nd to ^{150}Nd	-	5.078×10^{-4}

Figure 1



[Short/Long]
10^4
10^2
10^0
10^-2
10^-4
0
5
10
Atomic Number
Te
Xe
Sm
Ba
30
40
50
60
70
Atomic Number
Pu
90
100
110
Atomic Number

Figure 2



[Short/Long]
10^4
10^2
10^0
10^{-2}
10^{-4}
0
5
10
30
40
50
60
70
90
100
110
Atomic Number
Atomic Number
Atomic Number
Xe
Ba
Sm
Gd
Pu

Figure 3

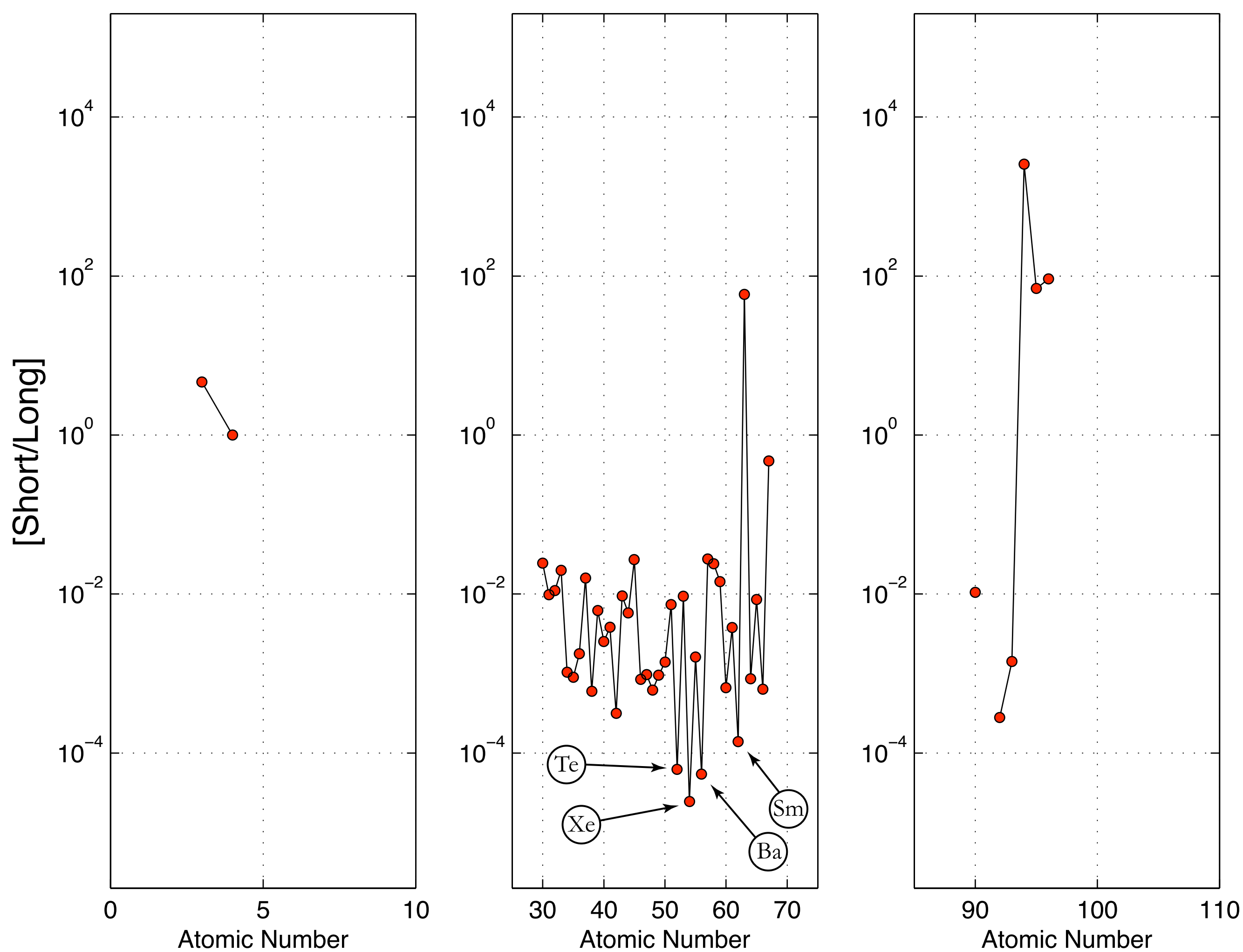

[Short/Long]
10^4
10^2
10^0
10^{-2}
10^{-4}
0
5
10
Atomic Number
30
40
50
60
70
Atomic Number
Te
Xe
Ba
Sm
90
100
110
Atomic Number

Figure 4

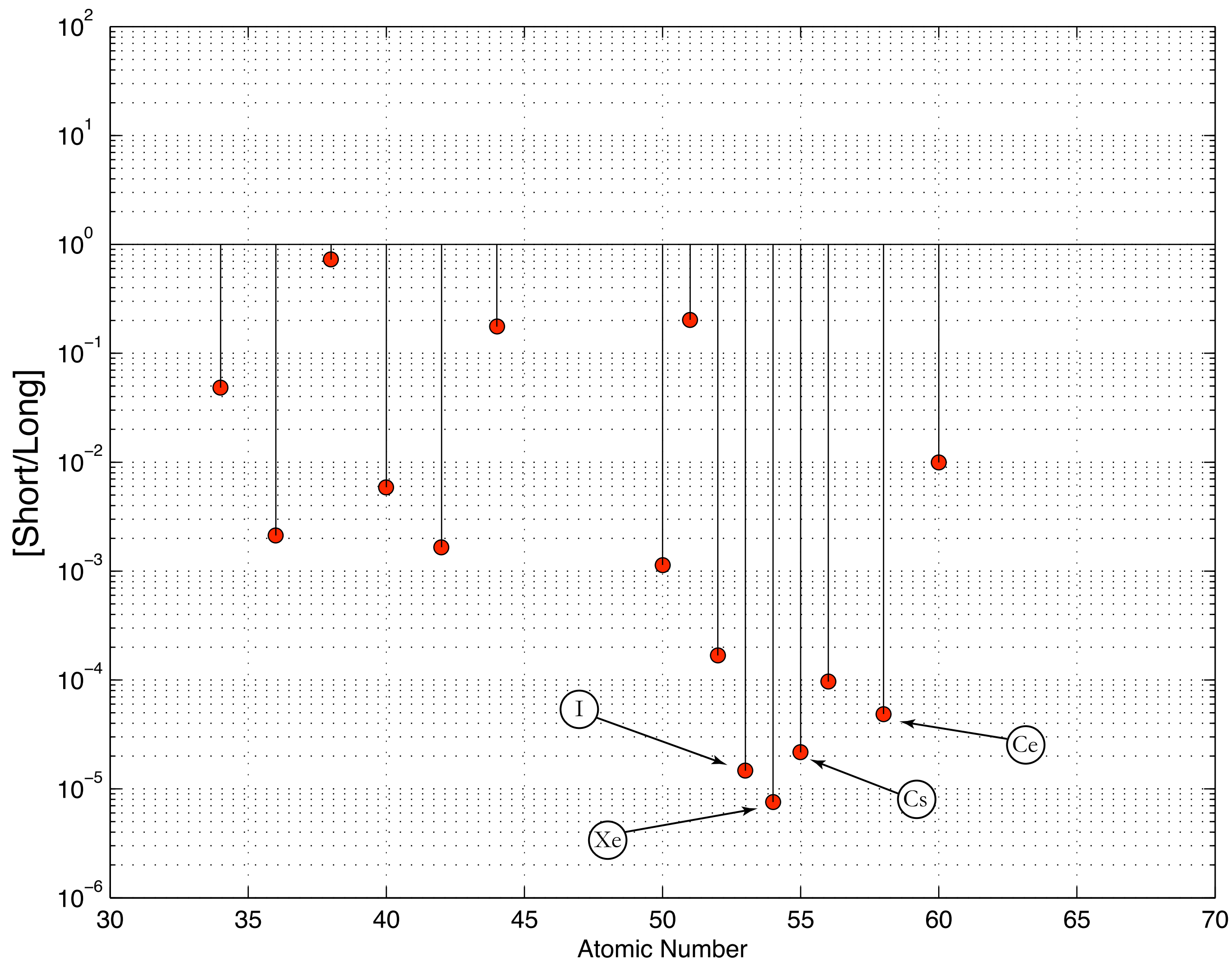
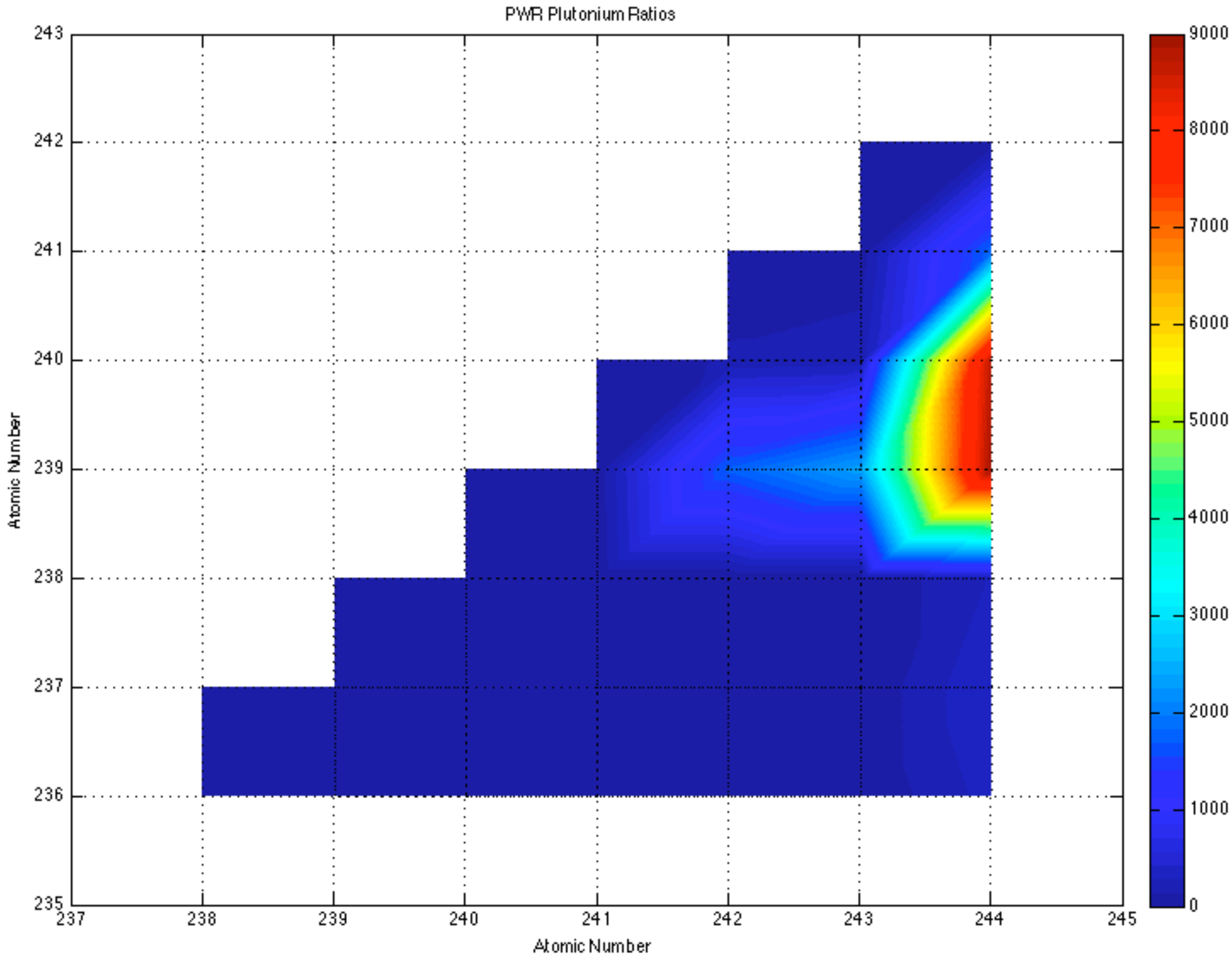


Figure 5


PWR Plutonium Ratios
Atomic Number
Atomic Number
235
236
237
238
239
240
241
242
243
237
238
239
240
241
242
243
244
245
0
1000
2000
3000
4000
5000
6000
7000
8000
9000